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SCINTILLATION SPECTROMETER SBL-1 FOR THE X-RAY DENSITOMETER OF RADIOACTIVE TECHNOLOGICAL SOLUTIONS

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Results of property study of the developed high-acting scintillation spectrometer to be used in x-ray densitometer of radioactive solutions with ^{241}Am “transmitting” radiant are presented. Fast crystals $YAlO_3$:Ce (Ø25x0.4 and Ø25x1.0 mm) are used as scintillators. Strong energy resolution dependence of the spectrometer with thin scintillators at crystal illumination (from 16.3 to 20.3 % for a γ-line 59.5 keV) is revealed. The spectrometer input count-rate is over $5{\cdot}10^5$ 1/s, and spectrum accumulation speed is not lower than $2{\cdot}10^5$ 1/s. Due to the conversion gain program stabilization the relative shift and the peak broadening E_γ = 59.5 keV do not exceed 0.25 % and 7.5 %, accordingly.

Keywords: $YAlO_3$:Ce, x-ray scintillation spectrometer, thin crystal, ^{241}Am, radioactive solutions, program stabilization, high count-rate, energy resolution.

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Introduction

Modern densitometers for industrial radiochemical applications are based on scintillation spectrometers with NaI(Tl) crystal. Radionuclide ^{241}Am (E_γ = 59.5 keV) is frequently used as a source of “transmission” radiation. Densitometric analysis of radioactive solutions sets the following requirements for the spectrometer used in the device:

1. Low detection efficiency for higher energy γ-ray (this is mainly radiation of radionuclide ^{137}Cs with energy E_γ =661.6 keV). Otherwise Compton continuum from scattering in the detector reduces “contrast” of density measurements. This problem is solved by an appropriate choice of corresponding scintillator thickness.
2. Good resolution on analytical line E_γ = 59.5 keV reduces the contribution to a statistical error caused by continuous allocation under the analytical peak. In scintillation γ-spectrometers the level of the resolving ability obtained almost completely depends on the detector material properties – light yield (*LY*) of scintillator measured in a number of light quantums per 1 keV of the absorbed γ-quantum energy and quantum efficiency (*QE*) of a photomultiplier photocathode. The product of these two values defines the statistical component of the resolution η_{stat} (LY_{NaI} = 36÷38 ph/keV; *QE* = 0.25÷0.43). In the x-ray energy range the light yield of most inorganic scintillators depends on the secondary electron energy [1]. The dependence and inhomogeneity in the crystal lead to the so-called intrinsic resolution η_{intr}, which contribution in the abovementioned energy range is comparable to η_{stat}.
3. While densitometrically measuring radioactive solutions, it is desirable to have the greatest possible activity of a “transmitting” radiant. Radiation resource of a scintillation crystal is therefore to be taken into account. NaI(Tl) crystals in this regard do not seem to be the best choice.
4. Spectrometer fast-action is critical in these applications, since the accuracy of the results depends mainly on it. The most important indicators of fast-action are the maximum input count-rate R_{i_max} and the maximum speed of spectrum accumulation R_{o_max}. R_{i_max} is defined as the maximum count-rate at which the relative broadening $\Delta\eta/\eta$ and shift $\Delta E/E$ of the analytical peak do not exceed the specified values. It is convenient that at $R_i = R_{i\_max}$ $\Delta\eta/\eta$ ≤ 10 ÷ 15% and $\Delta E/E$ ≤ 0.1 ÷ 0.25 %. The scintillation spectrometer fast-action is affected by scintillator decay time, gain stabilization system, structure and circuitry of the electronic spectrometer path.

The aim of the present paper is to develop and study scintillation spectrometer count-rate properties with scintillators more perspective than traditional NaI(Tl) ones.

The choice of the scintillator

Over 450 modifications of scintillation materials have been described the literature [2]. However, only about ten of them have the main characteristics not worse than those of classical NaI(Tl) and at least one property of the former is superior to those of the latter. Regarding the fact that not all ten perspective scintillators are employed in industrial production, the choice is unlikely to be rich. We have chosen $LaBr_3$:Ce (Ø18×3 mm manufactured by our strategic partner – RADICO company, Obninsk, Kaluga region, Russia) as well as $YAlO_3$:Ce (Ø25x0.4 mm and Ø25x1.0 mm produced by CRYTUR company, the Czech republic). In the world literature $YAlO_3$:Ce crystals are referred to as YAP:Ce. Table presents comparison of three crystals mentioned above as described in a number of papers.

As the analysis of the data in Table shows, the two crystals we have chosen ($LaBr_3$:Ce and $YAlO_3$:Ce) are quite suitable for detecting the line E_γ = 59.5 keV.

Table

Comparative analysis of three X-ray scintillators

Performance	Scintillator type		
	NaI(Tl)	$LaBr_3$:Ce	$YAlO_3$
Specific Gravity r, g/cm^3	3.67	5.10	5.35
Light Yield *LY*, ph/keV	38	63	≈ 20
Thermal Instability Coefficient of *LY*, %/^{0}C	- 0.75 in range (-30 ÷ +50) ^{0}C [3]	+ 0.01 [4]	+ 0.05 in range (-70 ÷ +125) ^{0}C [5]
Decay Time t_{LD}, ns	230	16	27
Wavelength of Max. Emission $l_{\mathbf{max}}$, nm	415	380	370
Refractive Index *n*	1.85	≈ 1.90	1.95
Energy Resolution on Lines:			
^{57}Fe $\eta_{14.4\ keV}$, %	22 [6]	≈ 22.5 [7]	38 [8]
^{241}Am $\eta_{59.5\ keV}$, %	12÷14 [9]	11.6 [10]	15÷16 [11]; 16.1 [12]
^{57}Co $\eta_{122.1\ keV}$, %	13 [13]	7 [13]	10.5 [12]

The spectrometer device

The design of scintillation spectrometer SBL-1 is shown schematically on Fig. 1, and the way it looks like is given on Fig. 2. The scintillation crystal sizes have been chosen on the basis of the required dimensions of the spectrometer detecting part and the energy of the γ-line to be recorded (E_γ = 59.5 keV). The above mentioned as well as the fact, that maximums of their emission spectra are in the range λ_{max} = 370-380 nm define photomultiplier type (**PMT**) which is HAMAMATSU R3998-100-02 with super bialkali photocathode Ø25 mm, providing $QE_{\lambda= 350\ nm}$ = 0.35 and $QE_{\lambda = 400\ nm}$ = 0.34.

The spectrometer electronic section is unified as much as possible with the same section of portable-stationary spectrometer STARK-01 [14]. Like all spectrometer channels developed in our study, it contains functional elements apart from linear amplifier (**LA**) and analog-to-digital converter (**ADC**). The structure and circuit implementations of the spectrometer electronic section under discussion provide extremely high fast-action and accuracy of the spectrometer. First of all it concerns the "digital" base-line stabilizer (**DBLS**) [15], realizing Noise Free Additional Pulse Shaping (**NFAPS**) principle [16, 17]. Fundamental differences of this type stabilizers consist in the fact that they do not introduce count-rate dependent noise [18], base-line stability practically does not depend on amplitudes, shapes and widthes T_w of spectrometer pulses up to $DF = R_i{\cdot}T_w \le 2$ (dead time up to 200 %). The design principles of pile-up inspector and dead time corrector are such [17] that their performances are coordinated with those of DBLS in terms of the range of amplitudes and maximal input count-rate. High count-rate limitations arise from the photomultiplier tube – the gain change due to the average current flow.

In STARK-01 this problem has been solved by means of light-emitting diode gain stabilization system. When the spectrometer is used as a part of the densitometer with monoline "transmitting" radiation source, the light-emitting diode system appears to be redundant. Due to the rigid fixation of a base-line position in this case, one can employ the algorithm of the program spectrum stabilization. The principle of program stabilization activity comes to the following. While spectrum accumulation being carried out, the analytical peak position (in this case it is a line of ^{241}Am, E_γ = 59.5 keV) is regularly checked by means of differential spectrum. The given spectrum is calculated as $SP(k{\cdot}t_c) - SP((k - 1){\cdot}t_c)$, where *SP* is the obtained spectrum, *k* is a current reading cycle of the spectrometer information in the computer, t_c is an interval between reading cycles. Thus, differential spectrum measuring time is defined as a time interval between readings of the information from the spectrometer and is only t_c = 500 ms. It allows one to gain information on change of analytical peak position (*APP*) without any delay. Differential spectrum processing occurs with each reading of the spectrometer information in the computer (one time per 500 ms). If the information is

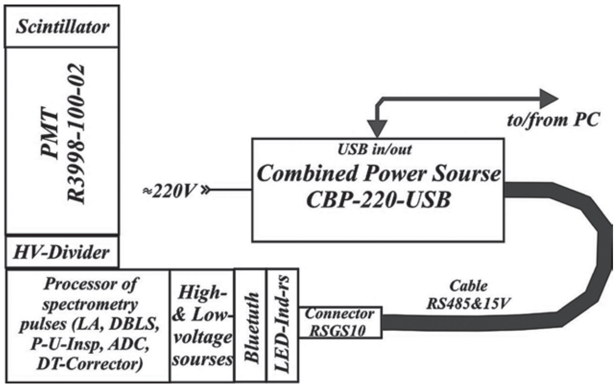

Fig. 1. Structure of *x*-ray scintillation spectrometer SBL-1

scarce (with count-rate lower than $2{\cdot}10^3$ 1/s), differential spectrum accumulates within several exchange cycles, otherwise APP definition error can make 0.2÷0.4 keV. The normalization coefficient in terms of the previous correction reflects E_γ(n) dependence declination change, where n is the number of the spectrometer channel. Initial spectrum normalized by energy is calculated according to the normalization coefficient. Our program correction algorithm of the spectrometer conversion coefficient drift eliminates the necessity in hardware fine adjustment of the amplification constant.

Experimental researches

The study began with YAP:Ce crystals of the sizes mentioned above. The crystals were placed on a thin Al-substrate with mirror surface at the crystal's side. The spectrometer scale calibration was carried out on two lines – 5.89 and 59.5 keV. The fact that the energy resolution $\eta_{59,5\ keV}$ strongly depends on whether

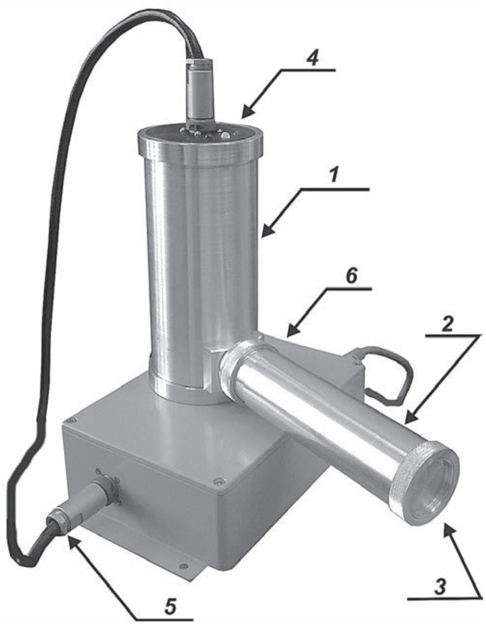

Fig. 2. Appearance of spectrometer SBL-1 with the exterior power supply: 1 - the case of an electronic section; 2 - the case of the PMT with a voltage divider and magnetic protection; 3 - an input window; 4 - light-emitting diode indicators; 5 - the combined communication cable; 6 - the combined power unit (KBP-220-USB)

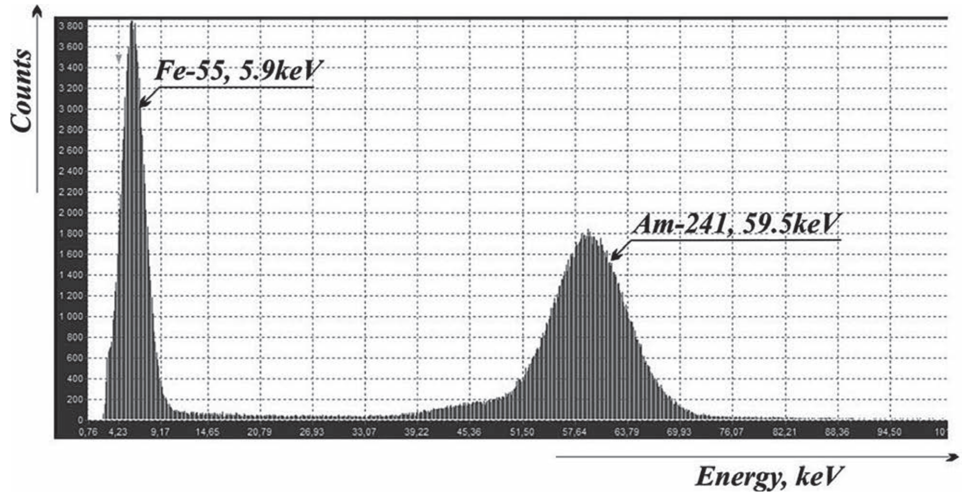

Fig. 3. ^{55}Fe + ^{241}Am spectrum. Exposure on YAP:Ce Ø25×1.0 mm crystal center through a collimator 3 mm. To the left from the peak E_γ = 59.5keV there is not allowed *Y* escape peak (E_γ = 59.5 - 14.96 keV)

a radiant ^{241}Am is collimated or not as well as whether it luminesces onto the crystal center or periphery was rather unexpected. Both crystals (Ø25x0.4 mm and Ø25x1.0 mm) showed the same result dependences on illuminating source position - $\eta_{59,5\ keV}$ = 18.3 % with the crystal center exposure through collimators Ø1.0mm and Ø3mm; $\eta_{59,5\ keV} \leq$ 20.0 % with the exposure at 1/4 of crystal diameter; $\eta_{59.5\ keV} \leq$ 19.9 % under conditions of wide geometry (the whole crystal is exposed).

The following procedures were carried out to eliminate this effect:

a) The lateral surface of crystals was wound up with a Teflon film for diffuse reflection. Improvements were not observed.

b) Aluminium substrates of crystals were removed. Measurements were performed with and without diffuse reflector around the crystal lapped to PMT. The diffuse reflector worsened the performances. The best results were obtained without the reflector:

– $\eta_{59.5\ keV}$ = 16.4 % at the crystal center exposure;

– $\eta_{59.5\ keV}$ ≤ 17.6 % with the exposure at 1/4 of crystal diameter;

– $\eta_{59.5\ keV}$ ≤ 20.4 % under the wide geometry conditions.

In all previous measurements the crystals were lapped to PMT employing optical silicone gel BICRON BC-630 (refractive coefficient *n* = 1.42) to make the change convenient. When the study was completed, crystal YAP:Ce Ø25x1.0 mm was pasted with two-component optical glue SKTN. Test measurements showed some resolution improvement (it is likely to have been caused by high value of glue refractive coefficient):

– $\eta_{59.5\ keV}$ =16.3 % at the crystal center exposure;

– $\eta_{59.5\ keV}$ ≤16.9 % with the exposure at 1/4 of crystal diameter;

– $\eta_{59.5\ keV}$ ≤20.3 % under the wide geometry conditions;

– $\eta_{59.5\ keV}$ ≤18.2 % under the wide geometry conditions after a thin lead collimator with a hole Ø20.0mm had been installed on the crystal.

Fig. 3 shows the measured ^{55}Fe + ^{241}Am spectrum at the exposure of the crystal Ø25x1.0 mm center. The source ^{241}Am luminesced through a steel substrate with ^{55}Fe agent, therefore there are no low energy lines of ^{241}Am in the spectrum. In the course of the studies interesting properties of PMT HAMAMATSU

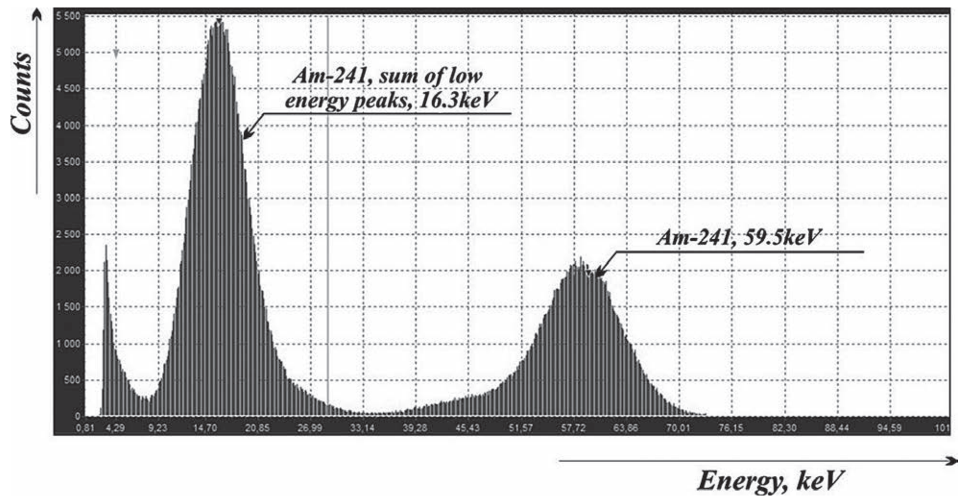

Fig. 4. ^{241}Am spectrum. YAP:Ce Ø25×1.0 mm crystal exposure under the “wide geometry” condition with an interior lead collimator Ø20.0 mm. The peak 16.3 keV - the total of not resolved low energy peaks ^{241}Am - earlier was killed by a steel substrate

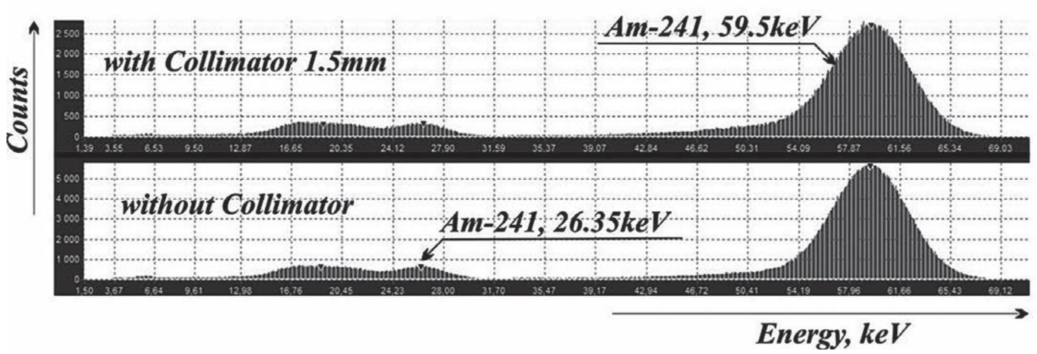

Fig. 5. [241]Am spectra obtained from $LaBr_3$:Ce crystal in two "geometries". The energy resolution practically does not depend on radiation collimation: $\eta_{59.5\ keV} \leq 11{,}0$ %; $\eta_{26.35\ keV} \leq 14.8$ %

R3998-100-02 came to light. Use of high voltage (*HV*) with the value close to that of nominal which is 1000 V gives rise to PMT autoemission. This leads to exponential distribution in the range of energies from zero to 10 keV with intensity up to 125 events per second. As high voltage decreases, intensity and amplitudes of the autoemission pulses drop. At *HV* = 800 V the mentioned intensity decreases down to 3÷5 1/s.

Fig. 4 shows [241]Am spectrum measured under "wide geometry" condition with interior Pb-collimator Ø20.0 mm.

The series of measurements on $LaBr_3$:Ce crystal showed that edge effects are not observed in this crystal packed with diffuse mirror unlike in YAP:Ce thin crystals. This is proved by the measured spectra (see Fig. 5). The absence of spatial sensitivity in this case might be caused by lower diameter to thickness ratio. This issue requires further studies.

The resolution considerably better than that of the previous crystals ($\eta_{59.5\ keV} \leq 11.0$ %) allows one to resolve peak 26.35 keV. Unfortunately, thick walls of the capsule which contained the scintillator made it impossible to study the spectrometer behavior at the lower energy region.

While experiments being made, the relative light yield of YAP:Ce crystals was measured as well. For this purpose the peak positions of [241]Am (E_γ = 59.5 keV) were measured in spectrometer SBL-1 adjusted on amplification to operate with $LaBr_3$:Ce. The peak positions relation expressed in channels was $N_{LaBr}/N_{YAP} = LY_{LaBr}/LY_{YAP} = 720/76 \sim 9.47$. Such estimation procedure is valid as zero values of the spectrometer scale expressed in energy units and channels practically coincide. If the statistics of the light quanta conversion in the scintillator were the primary resolution factor, energy resolutions would differ from each other by $9.47^{0.5}$, i.e. approximately by 3 times. The measured difference is only 16.3/11 ≈ 1.48. It testifies to the smaller contribution of the intrinsic resolution to YAP:Ce crystals in comparison with $LaBr_3$:Ce crystals which confirms to the data presented in the current literature.

The following can be said concerning high count-rate performances. As it was mentioned earlier, SBL-1 spectrometer electronic section is to the utmost unified with STARK-01 spectrometer section [14]. Decay times of YAP:Ce and $LaBr_3$:Ce crystals have the same order of magnitude (see Table). Due to this fact, the above-mentioned spectrometers show almost the same fast-action. The measured microscopic dead time T_D (time required for one event processing at high count-rates) is T_D = 1.725 μs for SBL-1. It guarantees the maximum input count-rate $R_{i\_max} \approx 5.75 \cdot 10^5$ 1/*s*. The maximum count-rate of free amplitude code pile-up is $R_{o\_max} \approx 2.1 \cdot 10^5$ 1/*s*. Such performance can be achieved with NaI(Tl) crystals, but only by incomplete integration of the light flashes [19]. This is equivalent to ≈ 16% decrease of the scintillator light yield, which leads to additional energy resolution degradation. It should be noted that the fast-action limitation of the spectrometer with YAP:Ce-scintillator is not due to the crystal properties, but to the electronic components used.

Conclusion

The designed spectrometer easily meets the requirements to be employed in the specific densitometric installation and can be further improved.

The fast-action achieved in this paper is not ultimate. At YAP:Ce crystal decay time τ_{LD} = 27 ns almost complete integration of the light flashes occurs within $T_{int} \geq 6.9\tau_{LD}$ = 186 ns. If spectrometric pulses with time to maximum (peak time) T_{peak} = 200 ns and the width time T_w = 600 ns are formed, as it was done in [20] for diamond detector signal, the spectrometer performance will be characterized by the values of the maximum input count-rate as well as spectra accumulation rate $R_{i\_max} \geq 1.5 \cdot 10^6$ 1/s and $R_{o\_max} \geq 5 \cdot 10^5$ 1/*s*, respectively. The principle of spectrometer electronic channel creation, however, remains unchanged.

Regarding the energy resolution, replacement of thin YAP:Ce crystal by $LaBr_3$:Ce one undoubtedly leads to the resolution increase (see Fig. 5). The challenge is that the scintillator production technology fails to allow one to reach thickness below 3 mm. This will lead to an almost five-fold increase in γ-radiation background download from [137]Cs contained in the samples under analysis.

YAP:Ce crystal has one more advantage over other crystals. The price of the former (≈500 €) is quite competitive with that of thin crystal NaI(Tl) currently in use, whereas vacuum-tight packaging for $LaBr_3$:Ce crystal makes it rather expensive.

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